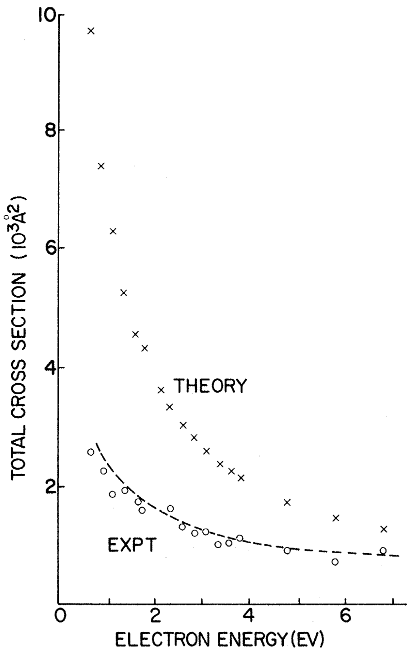

FIG. 3. Total cross sections: crosses, first Born approximation; circles, experimental fit with FBA; dashed curve, scaled maximum scattering out.

CsF.

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Pressure Shift of the Magnetic Resonance Line of Neon in a He-Ne Laser

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A pressure shift of the magnetic resonance line of the $3s_2$ level of neon in a He-Ne laser operating at 0.633-μm π transitions was observed by varying the partial pressure of helium. The shift in the resonant angular frequency was $(2.8 \pm 0.5) \times 10^7$ sec^{-1} Torr^{-1}, and the ratio a of the shift to the pressure broadening (half width at half-maximum) was 0.78 ± 0.14.

Pressure shifts of lines in the optical region have been reported by many authors, and such shifts have been explained primarily by the adiabatic collision theory, in which the atomic collisions change the atomic wave functions and do not induce transitions between the atomic energy levels. However, in the rf or microwave region, the mean kinetic energies of the colliding atoms exceed greatly the energy splitting, so that diabatic collisions have been considered to be important for the collisional broadening of the lines, and hence the pressure shifts have been expected to be very small. Although in the microwave region experiments have been reported on the shifts of the inversion spectrum of ammonia,[1,2] and on the hyperfine spectra of atomic hydrogen,[3,4] deuterium,[4] tritium,[4] sodium,[5] potasium,[6] rubidium,[7] and cesium,[8] the investigation of pressure shifts of lines in the rf region has not been reported.

In this Letter, we report on the experimental evidence of the pressure shift of the magnetic resonance line of the $3s_2$ level of neon in a He-Ne laser, which is caused by collisions with helium atoms. The laser operated at 0.633-μm π transitions on multilongitudinal modes of a cavity. The shift was $(2.8 \pm 0.5) \times 10^7$ sec^{-1} Torr^{-1}, while the resonant angular frequency was about 1.0×10^9/sec. The observation of the magnetic resonance of the laser level $3s_2$ was made by using the techniques of optical-rf double resonance, which have recently been applied to neon in He-Ne lasers operating at 1.15, 1.52,[9-11] and 0.633 μm,[12] but in these experiments pressure shifts of the magnetic resonance lines were not observed.

A detailed description of the present experimental apparatus was reported earlier.[12] The dc-excited laser in the transverse magnetic field H had a mirror spacing of 104 cm, which resulted in a frequency separation of about 144 MHz between axial modes; the Brewster-angle windows were oriented so that the electric vector of the laser field was parallel to the field H, i.e., only the π polarization state was allowed to oscillate. The laser field in the cavity has two roles: to create the alignment in the laser levels, and to monitor the magnetic resonances of these levels. The rf field H_1 perpendicular to the field H was applied by Lecher wires placed along the laser tube. In the present experiment, an improvement was made in the stability of the frequency of the rf field H_1. The frequency was kept constant to 158.4 MHz, which was the sixth harmonic of the original frequency of the stabilized crystal oscillator. The amplitude of the rf field H_1 was estimated as about 1.5 G from experimental results on the broadening of the resonance line due to the rf field. This value of H_1 was weak enough not to saturate the Zeeman transitions and to permit neglect of the Bloch-Siegert effect.[13] The static field H and the modulation field of 400 Hz were applied by a set of double Helmholtz coils,[14] the uniformity of which was a few parts in 10^4 over the discharge length, so that we could neglect the broadening of the line due to the field gradient in calculating the linewidth. The laser output was detected by a solar cell and applied to a lock-in amplifier, the output of which was displayed on an X-Y recorder as the static magnetic field H was swept through the resonance of about 87 G. We gave particular attention to reduction of the error in the measurement of the field H. The horizontal axis on the X-Y recorder was driven by the current to the double Helmholtz coils, and the value of H was always checked by a Hall magnetometer with an accuracy of 1%. We also measured the conversion ratio of the value of H to the current with an accuracy of 0.1% by means of an optically pumped Cs magnetometer, while applying a weak magnetic field of about 1 G.

It is known that the g factors of the $3s_2$ and $2p_4$ levels of neon are quite close to each other,[15] and thus the magnetic resonances of both laser levels are expected to occur simultaneously. However, our analysis, in which we applied the double-resonance theory of Culshaw[16,17] to the present case, showed that when we applied a weak rf field such as that in the present experiment, the resonance signal appearing as the intensity change of the laser output was mainly due to the magnetic resonance of the $3s_2$ level.

Observation of the resonance was made by varying the partial pressures of neon and helium independently. We used one laser tube, which was sealed off every time a particular partial pressure was studied. In order to avoid any systematic error, we varied these partial pressures in a random way, instead of in increasing or decreasing order.

Figure 1 shows the width $\Delta\omega$ (half width at half-maximum) and the center magnetic field H_0 of the resonance line as a function of the neon pressure P_{Ne}, while the helium pressure P_{He} was kept

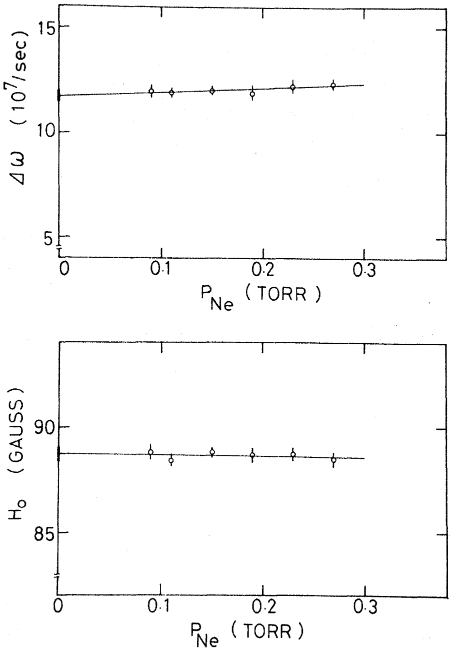

FIG. 1. Width $\Delta\omega$ and center magnetic field H_0 of the resonance line as a function of neon pressure P_{Ne}. The helium pressure P_{He} is kept constant at 0.9 Torr. The solid lines represent least-squares–fit values.

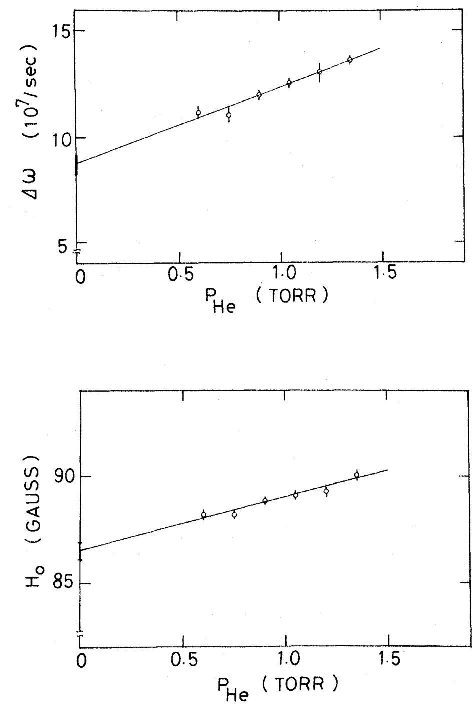

FIG. 2. Same as for Fig. 1 but for helium pressure P_{He}, with P_{Ne} kept constant at 0.15 Torr.

constant at 0.9 Torr. In the calculation of $\Delta\omega$, we assumed that the observed signal was the derivative of the Lorentz line shape, and we used a value for the g factor of 1.305 ± 0.005, which was obtained from the extrapolated value of H_0 to zero neon and helium pressures. In Fig. 1 we see a slight negative shift of H_0 with Ne pressure, but it is very small and is within our experimental error, so that we cannot discuss it further. Figure 2 shows the values of $\Delta\omega$ and H_0, where P_{He} is varied and P_{Ne} is fixed at 0.15 Torr. In this case we can see a relatively large positive shift of H_0 with He pressure. Summarizing these results, we can express the values of $\Delta\omega$ and H_0 as follows:

$$\Delta\omega = [(8.4 \pm 0.73) + (3.65 \pm 0.45)P_{\text{He}} + (2.04 \pm 1.42)P_{\text{Ne}}] \times 10^7/\text{sec},$$

$$H_0 = (86.5 \pm 0.4) + (2.49 \pm 0.40)P_{\text{He}} + (-0.55 \pm 1.5)P_{\text{Ne}}\ \text{G},$$

where P_{He} and P_{Ne} are in Torr. From the dependence on P_{He} and P_{Ne} of the linewidth $\Delta\omega$, we can easily obtain the alignment-destroying cross sections for the $3s_2$ level as $(9.5 \pm 1.1) \times 10^{-15}$ cm^2 for the collisions with helium atoms and $(13 \pm 9) \times 10^{-15}$ cm^2 for the collisions with neon atoms. It should also be noted that neither broadening nor shift of the resonance line was observed as we varied the discharge current which might be proportional to the electron density.

As described above, the experimentally determined ratio a of the shift to the broadening was 0.78 ± 0.14 for the collisions with helium atoms. It is very interesting that this value is quite close to the value of 0.726 predicted by the phase-shift theory of Foley[18] in which the interatomic interaction of colliding atoms is assumed to be the Van der Waals interaction, although this theory has been considered to be invalid for lines in the rf region where adiabatic collisions have been considered to be unimportant.

The collision cross sections for mixing of the Zeeman sublevels of the excited states caused by resonant collisions were calculated by D'yakonov and Perel'.[19] More recently some authors[20-23] have generalized the theory of resonant and nonresonant collisions of Byron and Foley[24] and Omont,[25] and derived the general expression for the cross sections for the relaxation of the multipole moment $\sigma^{(\varkappa)}$. In these theories, the interatomic interaction is assumed to be the electric dipole-dipole interaction for resonant collisions, and the Van der Waals interaction for nonresonant collisions. These theories predict the pressure shifts of the lines in the optical region but the shifts are independent of the magnetic quantum numbers of the related energy levels, which may result from the assumed interatomic interaction in the theories. Thus it seems very hard to explain the pressure shift obtained in the present experiment by use of these theories. In the present case, an additional interaction might be important, such as magnetic-type interactions. The collisional cross sections derived from the above theories have been in relatively good agreement with experiments except for experiments on collisions with helium atoms. Grossetete,[26] Barrat *et al.*,[27] and Faroux and Brossel[28] have found that a discrepancy exists between the experimental value of the disorientation cross section for the 6^3P_1 level of mercury perturbed by helium atoms and the theory in which the Van der Waals interaction is assumed. A similar discrepancy has also been found by Carrington and Corney[23] for the $2p$ levels of neon which are also perturbed by helium atoms. Carrington and Corney have suggested that this discrepancy might be due to the assumed interatomic interaction in which the repulsive part is ignored. However, it can also be considered that these discrepancies are closely related to the present experimental result, that collisions with helium atoms cause a relatively large pressure shift in the magnetic-resonance line of neon.

It is already known that collisions with helium atoms cause a shift of the 0.633-μm optical line of neon of about 12×10^7 sec^{-1} Torr^{-1} toward the blue.[29,30] As the $3s_2$ level is strongly perturbed by the laser field, we have to consider the process relating this shift of the optical line to the shift of the magnetic resonance line of the $3s_2$ level. One of the most probable processes might be the light shift[31] by the intense laser field. The light shift due to virtual transitions,[32] which has been observed in the magnetic resonance of the ground states of mercury[33] and of alkali atoms,[8,32] cannot cause the shift in the present case, since the laser field is in the π polarization state. On the other hand, the light shift due to real transitions[34] takes place when the g factors of the optically coupled levels are different and the light is in the π polarization state. However, this kind of shift is not important in the present case since the g factors of the $3s_2$ and $2p_4$ levels are approximately the same, and practically no shift of the resonance line could be observed when the laser intensity was varied.

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